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Optimization of direct conversion of methane to liquid fuels over Cu loaded W/ZSM-5 catalyst

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Abstract

The optimization of methane conversion to liquid fuels over copper loaded W/ZSM-5 catalyst was studied by utilizing experimental design from 'Statsoft Statistica' version 6.0 software. Response surface methodology was employed to determine the optimum methane conversion and C_5^+ selectivity. Numerical results indicated the optimum methane conversion of 29.4% with the corresponding C_5^+ selectivity of 57.2% were achieved at 12.3 vol% of O_2, 203.9 ml/min of total feed flow rate, and %W doped of 3.2 wt%. The optimum C_5^+ selectivity of 70.2% was attained at 7.6 vol% of O_2, 208.9 ml/min of total feed flow rate, and 3.2 wt% of W content with the corresponding methane conversion of 26.7%. By means of variance analysis and additional experiments, the adequacy of this model is confirmed.

Keywords: Optimization; Direct conversion; Methane; Liquid fuels

1. Introduction

Methane, the principal component of natural gas, can be converted to produce liquid fuels and chemicals of commercial importance. However, the commercialization of the direct conversion process remains a challenging goal as the engineering and chemistry involved are quite complex. In general, there are two routes for converting methane to liquid fuels: indirectly or/and directly. The indirect route is a two-step process whereby natural gas is first converted into synthesis gas (a mixture of H_2 and CO), and then into gasoline range. The direct route is a one step process in which the natural gas is reacted with oxygen (or another oxidizing species) to give the desired product directly.

Many researchers studied the applicability of HZSM-5 and modified ZSM-5 zeolite to the direct conversion of methane to liquid hydrocarbons [1–9], but the conversion and selectivity remained low making the process not lucrative economically. The direct partial oxidation of methane to liquid hydrocarbons was reported by Han et al. [1,2]. They found that liquid hydrocarbons could be produced from the reaction between CH_4 and O_2 over metal loaded ZSM-5. However, low conversion of methane was obtained due to a high formation of CO_x as side products. They concluded liquid hydrocarbons could be produced from the reaction between CH_4 and O_2 if CH_4 or C_2H_6 dehydrogenation and olefin oxidation functions of the metals in the metal–ZSM-5 catalysts are in balance. Different reactor configuration could also be used to achieve a better catalytic result as demonstrated by Pak et al. [9]. In their study, a high yield of liquid hydrocarbons (80%) was achieved using a two-reactor system with recycle, one for oxidative coupling of methane and the other for oligomerization reactor.

Previous studies on the conversion of methane to higher hydrocarbons generally accepted that CH_4 in the absence of O_2 reacted to form ethylene as an intermediate product on metal active sites by dehydrogenation, then followed by the formation of liquid hydrocarbons by oligomerization over acid sites of the catalyst. Recently, it has been shown that Mo supported on HZSM-5 are active and selective for conversion of methane to aromatics [10–17]. Wang et al. [16] studied the conversion of methane to aromatics over Mo/HZSM-5 based catalysts under non-oxidative condition and found that Mo species is transformed to Mo carbide species which is an active phase for methane transformation to higher hydrocarbons. Liu et al. [17] reported that methane

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